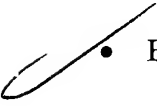


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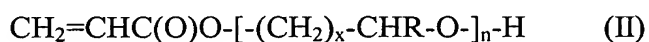
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**REMARKS**

Applicants respectfully request reconsideration of the application, as amended, in view of the following remarks.

The present invention as set forth in **amended Claim 1** relates to a method for preparing an **asymmetric (meth)acrylate crosslinking agent**

comprising **reacting an hydroxyacrylate** of formula (II)



**with methacrylic anhydride to form an asymmetric (meth)acrylate crosslinking agent** of formula (I) and methacrylic acid



wherein,

x = 1, 2, or 3,

R = H or CH<sub>3</sub>, and

n = 1-100;

**wherein a reaction product containing the asymmetric (meth)acrylate crosslinking agent comprises less than 2 wt.% of a diacrylate, dimethacrylate, or mixture thereof.**

The compound of formula (I) represents an asymmetric compound having an acrylate end group (CH<sub>2</sub>=CHC(O)O-) and a methacrylate end group (-C(O)CCH<sub>3</sub>=CH<sub>2</sub>).

As described in the specification under Discussion of the Related Art at pages 1 and 2:

“In addition, acryloyl chloride and methacryloyl chloride are also used for the synthesis of (meth)acrylate esters. In the case of acrylate esters in particular, however, **care must be taken for effective binding of liberated HCl, to prevent the formation of byproducts containing chlorine.**

Application No.: 10/042,232

Reply to Office Action of: October 1, 2003

...The **synthesis of methacrylate esters starting from methacrylic anhydride** and various alkanols with an additional ester function (lactate esters) **is distinctly less successful**. Despite catalysis with sulfuric acid, an excess of methacrylic anhydride and 5 hours of heating at 130 °C, conversion of only about 50 % is achieved. In addition, the **separation of unreacted methacrylic anhydride presents problems** (C. E. Rehberg et al., Journal of the American Chemical Society, Vol. 67, 210 (1945)).”

In contrast, to the known processes, the present invention provides an asymmetric cross-linking agent that has a high purity. In addition, known processes require the use of for example, large quantities of trialkylammonium chloride to trap liberated HCl when using acid chlorides. However, the present invention provides a method for producing asymmetric acrylic esters without additional purification steps.

None of JP64-87608 , Organic Chemistry, Mazur et al (US 5,149,642), Emmons (US 5,243,069), Matheisen et al (US 5,219,479), Thanawalla et al (US 4,618,703), Powanda et al (US 4,859,792) disclose or suggest to use methacrylic anhydride to form an asymmetric (meth)acrylate crosslinking agent or that the resulting product comprises less than 2 wt.% of a diacrylate, dimethacrylate, or mixture thereof, as claimed. The references use an acid chloride instead of acid anhydride as previously pointed out in the response filed August 7, 2003.

The Examiner has taken the position that “acid chlorides and acid anhydrides are equally and conventionally used in the art of organic chemistry for esterification.” (page 3 of the Office Action of October 1, 2003). However, an asymmetric cross-linking agent that has double bonds cannot be formed using an acid chloride without loss of monomer due to polymerization. In fact liberated HCL can add to the double bonds and cause polymerization, thereby decreasing the yield. However, this presents a problem, particularly in industrial processes in which the yield is an important factor in running a profitable reaction. The

separation of by-products is complicated, time consuming and expensive. On the other hand, as discussed above: "...The **synthesis of methacrylate esters starting from methacrylic anhydride** and various alkanols with an additional ester function (lactate esters) **is distinctly less successful.**" The conversion is only about 50% and unreacted anhydride must be separated (specification at page 1, last paragraph). Thus, one of ordinary skill in the art would not have a motivation or reasonable expectation of success to simply exchange acid chloride for acid anhydride because this may be less successful when the formation of double bond containing esters is involved. However, using the method of the present invention, high yields can be obtained as shown in the Example at page 9 of the specification (>90%).

Organic Chemistry at page 253 also describes what happens if the HCl being produced as a secondary product during conversion to the ester is not absorbed: "it might add to carbon-carbon double bond if one were present elsewhere in the molecule." This, however, is precisely the case here. **As noted above, the compound of formula (I) represents an asymmetric compound having an acrylate end group ( $\text{CH}_2=\text{CHC}(\text{O})\text{O}-$ ) and a methacrylate end group ( $-\text{C}(\text{O})\text{CCH}_3=\text{CH}_2$ ).** In contrast to methacrylic acid esters, acrylic acid esters are highly active vinologous carbonyl compounds, which add on free HCl almost quantitatively at temperatures as low as  $-10^\circ\text{C}$ . See Organic Chemistry, p. 569, General working procedure for the addition of hydrogen halide to vinologous carbonyl compounds, attached herewith. The last column of the Table shows a high yield of 80 % for the addition of HCL to acrylic acid ethyl ester.

This secondary reaction of producing HCl does not exist during the use of methacrylic acid anhydride. Thus, the claimed method for synthesis of asymmetric acrylic acid esters

Application No.: 10/042,232  
Reply to Office Action of: October 1, 2003

works without complex removal of large quantities of trialkylammonium chloride by filtration or washing processes. Trialkylammonium chloride is needed to trap HCl when using acid chlorides.

In the claimed method, only methacrylic acid, which can be easily distilled off from the mixture, is produced as secondary product.

It must be pointed out once again that the claimed method for synthesis of asymmetric acrylate esters works without the slightest contamination by lachrymatory chlorine-containing secondary products.

Therefore, the rejection of Claims 1-13 and 18 under 35 U.S.C. §103(a) as being unpatentable over JP64-87608 in view of “Organic Chemistry” by John McMurry, or separately in view of any one of the following Mazur et al (US 5,149,642) or Emmons (US 5,243,069) or Matheisen et al (US 5,219,479) or Thanawalla et al (US 4,618,703) or Powanda et al (US 4,859,792) is believed to be unsustainable as the present invention is neither anticipated nor obvious and withdrawal of this rejection is respectfully requested.

In addition, the rejection of Claims 1, 2, 5-10, 12, and 13 under 35 U.S.C. §103(a) as being unpatentable over SU 630249 in view of “Organic Chemistry” by John McMurry or separately in view of any one of the following Mazur et al (US 5,149,642) or Emmons (US 5,243,069) or Matheisen et al (US 5,219,479) or Thanawalla et al (US 4,618,703) or Powanda et al (US 4,859,792) is respectfully traversed.

None of the cited references disclose or suggest a method as claimed.

SU 630249 also uses acid chlorides instead of the claimed methacrylic anhydride. The secondary references do not cure the defects of the primary reference. There is no

Application No.: 10/042,232

Reply to Office Action of: October 1, 2003

reasonable expectation of success when using acid anhydrides in combination with polymerizable alcohols. As discussed in the specification and cited above, it is known that synthesis of methacrylate esters starting from methacrylic anhydride is distinctly less successful. Thus, there is no motivation to use methacrylic anhydride to form an asymmetric (meth)acrylate crosslinking agent. Thus, even a combination of SU 630249, Organic Chemistry, Mazur et al (US 5,149,642), Emmons (US 5,243,069), Matheisen et al (US 5,219,479), Thanawalla et al (US 4,618,703), Powanda et al (US 4,859,792) does not result in the present invention.

Therefore, the rejection of Claims 1, 2, 5-10, 12, and 13 under 35 U.S.C. §103(a) as being unpatentable over SU 630249 in view of “Organic Chemistry” by John McMurry or separately in view of any one of the following Mazur et al (US 5,149,642) or Emmons (US 5,243,069) or Matheisen et al (US 5,219,479) or Thanawalla et al (US 4,618,703) or Powanda et al (US 4,859,792) is believed to be unsustainable as the present invention is neither anticipated nor obvious and withdrawal of this rejection is respectfully requested.

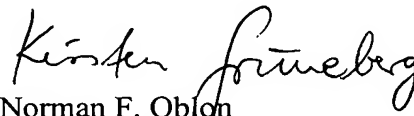
Applicants respectfully request that the Examiner acknowledge that the references cited in the **Information Disclosure Statement**, filed in the above-identified application on **November 26, 2003**, have been considered. For the Examiner's convenience a copy of Form PTO 1449 as filed on **November 26, 2003**, is attached herewith.

Application No.: 10/042,232  
Reply to Office Action of: October 1, 2003

Applicants submit that the present application is now in condition for allowance and early notice of such action is earnestly solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,  
MAIER & NEUSTADT, P.C.

A handwritten signature in black ink, appearing to read "Norman F. Oblon".

Norman F. Oblon  
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Kirsten A. Grueneberg, Ph.D.  
Registration No.: 47,297

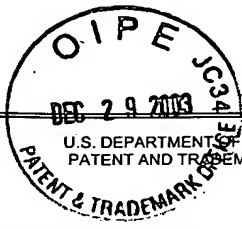
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Form PTO 1449  
(Modified)U.S. DEPARTMENT OF COMMERCE  
PATENT AND TRADEMARK OFFICE

ATTY DOCKET NO.

215503US0

SERIAL NO.

10/042,232

## LIST OF REFERENCES CITED BY APPLICANT

APPLICANT

Werner SIOL

FILING DATE

January 11, 2002

GROUP

1713

## U.S. PATENT DOCUMENTS

EXAMINER INITIAL		DOCUMENT NUMBER	DATE	NAME	CLASS	SUB CLASS	FILING DATE IF APPROPRIATE
	AA						
	AB						
	AC						
	AD						
	AE						
	AF						
	AG						
	AH						
	AI						
	AJ						
	AK						
	AL						
	AM						
	AN						

## FOREIGN PATENT DOCUMENTS

		DOCUMENT NUMBER	DATE	COUNTRY	TRANSLATION	
					YES	NO
	AO	0 899 286	03/03/99	Europe		
	AP					
	AQ					
	AR					
	AS					
	AT					
	AU					
	AV					

## OTHER REFERENCES (Including Author, Title, Date, Pertinent Pages, etc.)

	AW	Patent Abstracts of Japan; Vol. 012, No. 293, July 6, 1989; & JP 01 087608, March 31, 1989	
	AX	Patent Abstracts of Japan; Vol. 007, No. 108, May 11, 1983; & JP 58 029744, February 22, 1983	
	AY	Database WPI Section Ch, Week 197932, Derwent Publications Ltd., London, GB; Class A41, AN 1979-59492B XP002258093 & SU 630 249 A (AS USSR HIGH MOLECU), September 13, 1978	
	AZ	Yasukazu SAIMI et al; "Preparation and Visible Light Polymerization of Triethyleneglycol Acrylate Methacrylate"; Polymer Journal, Vol. 24, No. 4, pp. 357-363; 1992	<input type="checkbox"/> Additional References sheet(s) attached

Examiner

Date Considered

\*Examiner: Initial if reference is considered, whether or not citation is in conformance with MPEP 609; Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

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## 7.5. Literaturhinweise

569

## Allgemeine Arbeitsvorschrift für die Addition von Halogenwasserstoff an vinyloge Carbonylverbindungen

Man leitet unter Feuchtigkeitsausschluß trockenen Halogenwasserstoff in 0,2 Mol der frisch destillierten vinylogen Carbonylverbindung, die in Eis-Kochsalz-Mischung auf etwa  $-10^{\circ}\text{C}$  abgekühlt wird, mit einer solchen Geschwindigkeit ein, daß die Temperatur im Kolben nicht über  $-5^{\circ}\text{C}$  ansteigt. Nach Aufnahme der theoretischen Gasmenge (Gewichtskontrolle!) läßt man den verschlossenen Kolben bei  $0^{\circ}\text{C}$  über Nacht stehen. Das Reaktionsgemisch wird nacheinander mit Wasser, 10%iger Natriumhydrogencarbonatlösung und nochmals mit Wasser gewaschen, über Magnesiumsulfat getrocknet und destilliert.

Tabelle 7.239

Addition von Halogenwasserstoff an vinyloge Carbonylverbindungen

Endprodukt	Ausgangsprodukte	Physikal. Konstanten	Ausb. [%]
$\beta$ -Chlorpropionitril	Acrylnitril Chlorwasserstoff	$Kp_{-25}$ $87^{\circ}\text{C}$ , $n_D^{20}$ 1,4380	95
$\beta$ -Brompropionitril	Acrylnitril Bromwasserstoff	$Kp_{-25}$ $92^{\circ}\text{C}$ , $n_D^{20}$ 1,4789	90
$\beta$ -Chlorpropionsäure- äthylester	Acrylsäureäthylester Chlorwasserstoff	$Kp_{-25}$ $80^{\circ}\text{C}$ , $n_D^{20}$ 1,4254	80
$\beta$ -Brompropionsäure- methylester	Acrylsäuremethylester Bromwasserstoff	$Kp_{-15}$ $65^{\circ}\text{C}$ , $n_D^{20}$ 1,4542	80
$\beta$ -Brompropionsäure- äthylester	Acrylsäureäthylester Bromwasserstoff	$Kp_{-15}$ $78^{\circ}\text{C}$ , $n_D^{20}$ 1,4669	90
$\beta$ -Bromisobuttersäure- methylester	Methacrylsäuremethylester Bromwasserstoff	$Kp_{-25}$ $78^{\circ}\text{C}$ , $n_D^{20}$ 1,4551	80

## 7.5. Literaturhinweise

Darstellung von Acetalen, Mercaptalen, Azomethinen, Oximen, Hydrazonen und Hydrogensulfataddukten

O. BAYEN, in Houben-Weyl, Bd. VII/1, S. 413—488 (1954).

Darstellung und Reaktionen von Iminen

R. W. LAYNE, Chem. Reviews 63, 489—510 (1963).

Darstellung und Reaktionen von Enaminen

J. SZKUSZKOWICZ, Advances org. Chem. 4, 1—112 (1963).

# Organikum

ORGANISCH-CHEMISCHES GRUNDPRAKTIKUM

10. AUFLAGE



VEB DEUTSCHER VERLAG DER WISSENSCHAFTEN  
BERLIN 1971